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Intra-magnetoexciton transitions in semiconductor quantum wells

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ABSTRACT

Highly sensitive optically detected resonance experiments have shown that magnetoexcitons in GaAs-(Ga,Al)As semiconductor quantum wells have discrete internal energy levels, with transition energies found in the far-infrared (terahertz) region. Here we are concerned with a theoretical study of the terahertz transitions of light-hole and heavy-hole confined magnetoexcitons in GaAs-(Ga,Al)As quantum wells, under a magnetic field applied in the growth direction of the semiconductor heterostructure. The various magnetoexciton states are obtained in the effective-mass approximation by expanding the corresponding exciton-envelope wave functions in terms of appropriate Gaussian functions. The electron and hole cyclotron resonances and intra-magnetoexciton transitions are theoretically studied by exciting the allowed electron, hole and internal magnetoexcitonic transitions with far-infrared radiation. Theoretical results are obtained for both the intra-magnetoexciton transition energies and oscillator strengths associated with excitations from 1s - like to 2s, $2p_{\pm}$, and $3p_{\pm}$ - like magnetoexciton states, and from $2p_{-}$ to 2s – like exciton states. Present results are in overall agreement with available optically detected resonance measurements and clarifies a number of queries in previous theoretical work.

INTRODUCTION

The study of the optical properties of semiconductor heterostructures, such as GaAs-(Ga,Al)As quantum wells (QWs) and multiple quantum wells (MQWs), provides worthy information on the physical nature of confined electrons, holes, and Coulomb-bound states such as impurities and excitons. It is well known that excitons essentially dominate the optical properties of semiconductor heterostructures and, in particular, an external perturbation such as an applied magnetic field perpendicular to the GaAs and $Ga_{1-x}Al_xAs$ semiconductor layers is a powerful tool which is expected to provide valuable information on carrier subbands and exciton states via magneto-optical studies. Confined excitons in GaAs-$Ga_{1-x}Al_xAs$ QWs and MQWs under magnetic fields in the growth direction reveal themselves as a series of hydrogenic-like ground and excited magnetoexciton states, with the internal transition energies among the various exciton states in the far-infrared region (FIR - of the order of 10 meV or 2.4 THz). In particular, Salib et al [1] recently observed several internal excitonic transitions and found the 1s $\rightarrow 2p_+$ heavy-hole (*hh*) exciton transition as dominant in GaAs-$Ga_{0.7}Al_{0.3}As$ MQWs of L_w = 80 Å and L_w = 125 Å of well width. They have also assigned a "weak" feature and a "very weak" feature in the experimental spectra to *hh* magnetoexciton 1s $\rightarrow 3p_+$ and 1s $\rightarrow 4p_+$ transitions, respectively, based primarily on the magnitude of the energy separations with respect to the 1s $\rightarrow 2p_+$ transition. Also, some features present in the optically detected resonance (ODR) spectra by Salib et al [1] were attributed to hole cyclotron resonances (CRs) and others termed as of uncertain origin. In a simultaneous and independent experiment, Cerne et al [2] monitored changes in the excitonic PL that are induced by FIR radiation with the electric field polarized in the plane of the QW, and observed resonant FIR absorption by confined magnetoexcitons in GaAs - $Ga_{1-x}Al_xAs$ QWs under magnetic fields applied perpendicular to the well interfaces. The dominant resonance was assigned to the 1s $\rightarrow$ $2p_+$ *hh* exciton transition, and was found to persist even when the FIR electric field was comparable to the electric field which binds the exciton. More recently, Nickel et al [3] have used ODR spectroscopy to study electron and hole CRs and various internal excitonic transitions in a number of GaAs-(Ga,Al)As MQW structures. They concluded that more work is necessary to confirm the $2p_\pm$ assignments, to resolve the nature of the higher energy intraexcitonic transitions, and to observe light-hole (*lh*) CR and associated intraexcitonic transitions. From the theoretical point of view, Duque et al [4] have performed a study of 1s $\rightarrow$ $2p_\pm$, 1s $\rightarrow$ $3p_\pm$, and 1s $\rightarrow$ $4p_\pm$ *lh* and *hh* magnetoexcitonic transition energies in GaAs-$Ga_{1-x}Al_xAs$ QWs within a variational procedure in the effective-mass approximation, and although some of the theoretical magnetoexciton transition energies agree quite well with experimental measurements, other calculated results only reproduce qualitative features of experiment. In order to provide a better understanding of the terahertz transitions of confined magnetoexcitons in GaAs - $Ga_{1-x}Al_xAs$ QWs and of the ODR experimental data by Salib et al [1], Cerne et al [2], and Nickel et al [3], here we perform a more detailed theoretical study of the various intraexcitonic transitions in GaAs - $Ga_{1-x}Al_xAs$ QWs under magnetic fields applied along the QW growth direction.

THEORETICAL FRAMEWORK

We work in the effective-mass approximation and are interested in Wannier-exciton states in GaAs-$Ga_{1-x}Al_xAs$ QWs of width L_w in the presence of a magnetic field parallel to the growth direction of the heterostructure. We assume the spin-orbit splitting to be large enough that the interaction between J = 3/2 and J = 1/2 states may be disregarded. For simplicity, we take the relative motion of the carriers and that of the center of mass as independent, although one may only make this separation in the plane of the well [6], and write the exciton envelope wave functions as $F^{i}_{J^e_z,J^h_z}(\vec{r}_e,\vec{r}_h) \equiv F^{i}_{J^e_z,J^h_z}(\vec{\rho},z_e,z_h)$, where $\vec{\rho}$ is the *e-h* relative coordinate in the plane of the QW. In what follows, we will restrict our attention to independent excitons and discard the off-diagonal elements in the hole Hamiltonian [5], i.e., we neglect effects due to hole-subband mixing in the calculation. Image-charge effects are not considered and the *e-h* Coulomb interaction is assumed to be screened by an average static dielectric constant of the GaAs and $Ga_{1-x}Al_xAs$ bulk materials. The values of the square potential-well barriers $V_c(z_e)$ and $V_v(z_h)$ are determined from the Al concentration and assumed to be 65% and 35% of the total energy-band-gap discontinuity, respectively. Also, although actual measurements are for GaAs-$Ga_{0.7}Al_{0.3}As$ superlattices (SLs), we have ignored SL tunneling effects and performed calculations for single isolated GaAs-$Ga_{0.7}Al_{0.3}As$ QWs.

We may now expand the exciton envelope wave functions $F^{i}_{J^e_z,J^h_z}(\vec{\rho},z_e,z_h)$ in terms of single-particle $f^{J^e_z}_{n}(z_e)$ and $f^{J^h_z}_{m}(z_h)$ solutions of the effective-mass equation for electron or hole motion, respectively, along the z-axis of the QW, and write [7]

$$F^{i}_{J^e_z,J^h_z}(\vec{\rho},z_e,z_h)=\sum_{n,m}\psi^{J^e_z,J^h_z,i}_{n,m}(\rho,\phi)\, f^{J^e_z}_{n}(z_e)\, f^{J^h_z}_{m}(z_h), \qquad (1)$$

with

$$\psi^{J^e_z,J^h_z,i}_{n,m}(\rho,\phi)=\sum_{j}C^{J^e_z,J^h_z,i}_{n,m,j}\;\rho^{|s|}\,e^{is\phi}\,e^{-\frac{\rho^2}{\lambda_j^2}}, \qquad (2)$$

where the expansion is made in a restricted set of Gaussian functions with appropriate length parameters λ_j, and s is an integer associated with the conserved z component of the total angular momentum [7]. We assume the *hh* and *lh* exciton Hamiltonians of ref. [5], and the GaAs conduction-band effective-mass and dielectric constant as $m_e = 0.0665$ (in units of the free electron mass m_0) and $\varepsilon = 12.5$, respectively; the relevant mass parameters and the Luttinger valence-band parameters are taken from Bauer and Ando [5]. In what follows, magneto-exciton energy states are labeled as $n\ell m\,(J^e_z,J^h_z)$ which correspond to an $n\ell m$ -like exciton state composed of a J^e_z electron (with $J^e_z = \pm 1/2$) and a J^h_z hole (with $J^h_z = \pm 1/2, \pm 3/2$).

We also note that, in the dipole approximation, the $\alpha(\omega)$ magneto-absorption coefficient for the intraexcitonic 1s $\rightarrow$ np$_\pm$ transitions is essentially given by

$$\alpha(\omega)\propto\frac{1}{\omega}\sum_{f}\left|\left\langle F^{f}_{J^e_z,J^h_z}(\vec{\rho},z_e,z_h)\right|\,\hat{\varepsilon}.\vec{\mathbf{P}}_r\left|F^{1s}_{J^e_z,J^h_z}(\vec{\rho},z_e,z_h)\right\rangle\right|^2\,\delta(E_f-E_{1s}-\hbar\omega), \qquad (3)$$

where $\hat{\varepsilon}$ corresponds to the photon polarization and $\vec{\mathbf{P}}_r$ to the relative mechanical momentum of the *e-h* pair.

RESULTS AND DISCUSSION

We first point out that, in the calculation by Duque et al [4], the exciton envelope wave function is described as a product of variational hydrogenic-like wave functions and electron and hole **ground-state** solutions of the effective-mass equation for motion along the z-axis of the QW. In the present approach, the exciton wave function is written in terms of products of Gaussian functions with appropriate hole and electron single-particle states [contribution of the **ground state and excited states**, cf. eq. (1)]. For allowed spin-conserving transitions involving the lowest-energy exciton states, i.e., for the *hh* 1s $\rightarrow$ 2p$_\pm$ magnetoexciton transitions, one finds very good agreement between the two calculations, whereas for higher-energy *hh* 1s $\rightarrow$ 3p$_\pm$ and *lh* 1s $\rightarrow$ 2p$_\pm$ and 1s $\rightarrow$ 3p$_\pm$ magnetoexciton transitions both theoretical approaches end up in quantitatively different results. Of course, the present scheme is more reliable from the quantitative point of view, and better describes higher-energy states as the exciton envelope wave function in eq. (1) includes the effects of excited electron and hole single-particle in its expansion.

The energies corresponding to *lh* and *hh* 1s $\rightarrow$ 2p$_\pm$ and 1s $\rightarrow$ 3p$_\pm$ magnetoexciton transitions are shown in Fig. 1(a) for a 80 Å GaAs-$Ga_{0.70}Al_{0.30}As$ QW. Notice that the experimental FIR data by Salib et al [1] are in fair agreement with intraexcitonic theoretical transitions, although any assignment of the experimental higher energy FIR data to specific magnetoexciton transitions is difficult to make. Figure 1(b) displays the calculated *lh* and *hh* intraexcitonic magneto-absorption coefficient, for left- and right-circularly polarized light in the well plane, also in the case of a L_w = 80 Å GaAs-$Ga_{0.7}Al_{0.3}$As QW. It is then clear that the oscillator strength of the *lh* and *hh* 1s → $2p_\pm$ intraexcitonic transitions are of the same order of magnitude and that the *lh* 1s → $2p_\pm$ transitions should therefore be experimentally observable. The calculated results in Fig. 1(b) unambiguously indicate that, if one performs the experiment with appropriately polarized photons, both the *lh* and *hh* 1s → $2p_-$ exciton transitions should be noticeable in the measured spectra. Also, one clearly sees [for B = 1, 2, and 3 T in Fig. 1(b)] that weaker, higher energy features in $\alpha(\omega)$ corresponding to *lh* and *hh* 1s → $3p_\pm$ transitions should be observable, provided one is able to perform the experiment with higher spectral resolution; in fact, one notices that some of these higher energy transitions do show up in the experiment corresponding to the L_w = 80 Å sample [see Fig. 1(a)].

Although not shown here, we have evaluated the *lh* and *hh* 1s → $2p_\pm$ magnetoexciton transition energies and magneto-absorption coefficient for a 100 Å GaAs-$Ga_{0.70}Al_{0.30}$As QW. Results are in quantitative agreement with FIR data and, in particular, some higher-energy experimental transitions, which Cerne et al [2] do not assign to any specific intraexcitonic transition, are found to correspond to *lh* 1s → $2p_+$ magnetoexciton transitions. We have also calculated the *hh*1s – 2s dispersion (separation of 60 cm^{-1} at zero magnetic field), and it agrees quite well with the 1s – 2s energy separation obtained by Cerne et al [2] with PL and PLE measurements. Moreover, we found that the lowest FIR frequency resonance is due to free-electron CR and **not** to *hh* $2p_-$ → 2s transition energies, as suspected by Cerne et al [2].

In order to analyze the recent magnetoexciton experimental FIR data by Nickel et al [3], one should notice that, for wide wells, heavy and light holes should be strongly mixed, and hole-subband mixing must be taken into account. In order to mimic this effect, one may choose to continue to work with the parabolic one-particle Hamiltonian and adequately change the effective-mass parameters and dielectric constant in order to fit the FIR experimental transitions. Corresponding calculated results for the 1s → $2p_\pm$ and 1s → $3p_\pm$ magnetoexciton transition energies and magneto-absorption coefficient are shown in Fig. 2 for the L_w = 150 Å sample, with ε(GaAs) chosen as 13.9, and the conduction - electron effective mass chosen as 0.075 m_o. In Fig. 2(a), the full down-triangles magnetic-field dependent experimental FIR energies correspond to 1s → $2p_-$ calculated magnetoexciton transitions for a valence-band effective mass of 0.3 m_o, whereas the full up-triangles FIR data correspond to the 1s → $2p_-$ transitions for a valence-band effective masses chosen as 0.79 m_o. The value of 0.79 m_o for the valence-band in the case of L_w = 150 Å is certainly peculiar, but it reflects the unexpected experimental behavior [3], i.e., in the case of the L_w = 150 Å sample the difference of the 1s → $2p_-$ experimental magnetoexciton transitions (up- and down-triangles) is higher than in the L_w = 200 Å case - see data in ref. [3] – in which the mixing of *hh* and *lh* should be stronger. Further studies are necessary to clarify this point.

Summing up, theoretical results in GaAs-(Ga,Al)As QWs are obtained for intra-magnetoexciton transition energies corresponding to excitations from 1s - like to 2s -, $2p_\pm$ -, and $3p_\pm$ - like magnetoexciton states, and from $2p_-$ - to 2s – like states. We have also presented results for the $\alpha(\omega)$ magneto-absorption coefficient corresponding to the intraexcitonic 1s → $np_\pm$ transitions, in the dipole approximation, for the case of left- and right-circularly polarized photons. Finally, we have compared the present theoretical results with available optically detected resonance measurements, and obtained good overall agreement. Nevertheless, there are still several aspects of the magnetoexciton problem that are unclear or unexpected, and quite

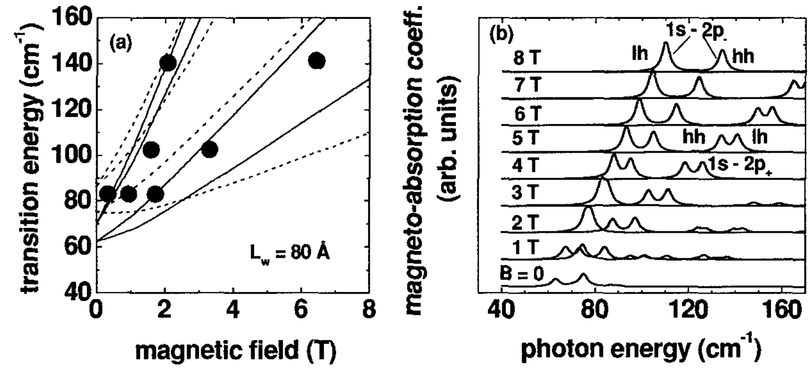

Figure 1. (a) Light-hole (dashed curves) and heavy-hole (full curves) 1s $\rightarrow$ 2p$_\pm$ and 1s $\rightarrow$ 3p$_\pm$ calculated magnetoexciton transition energies, with experimental data (full circles) taken from Salib et al [1]; (b) Intraexcitonic light-hole (*lh*) and heavy-hole (*hh*) 1s $\rightarrow$ np$_\pm$ magneto-absorption coefficient, for the case of left- and right-circularly polarized light in the well plane; the column of numbers on the left gives values of the applied magnetic field in Teslas (T). Results are for L_w = 80 Å GaAs-Ga$_{0.7}$Al$_{0.3}$As QWs under magnetic fields applied along the growth direction of the heterostructure.

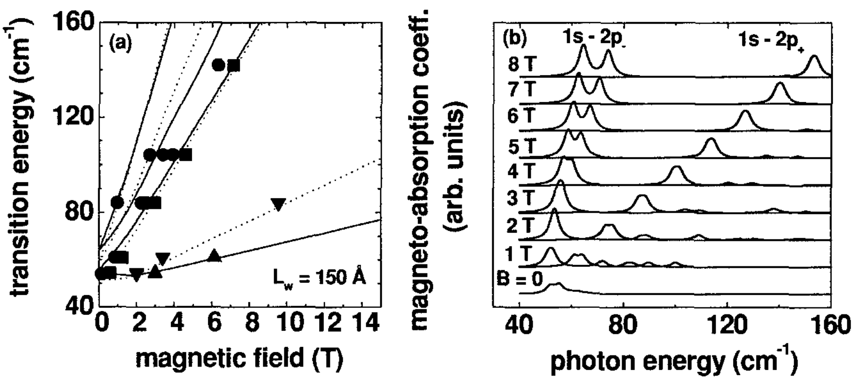

Figure 2. (a) Calculated 1s $\rightarrow$ 2p$_\pm$ and 1s $\rightarrow$ 3p$_\pm$ magnetoexciton transition energies, with experimental data (solid dots) taken from Nickel et al [3]. Calculations assume the in-plane heavy-hole mass as 0.79 m$_o$ (full curve) and 0.30 m$_o$ (dotted line); (b) Intraexcitonic 1s $\rightarrow$ np$_\pm$ magneto-absorption coefficient, for the case of left- and right-circularly polarized light in the well plane; the column on the left gives values of the applied magnetic field in Teslas (T). Results are for L_w = 150 Å GaAs-Ga$_{0.85}$Al$_{0.15}$As QWs under magnetic fields applied along the growth direction of the heterostructure.

certainly, further experimental and theoretical studies are needed. In that sense, it would be of interest to perform experimental studies with right-, left- and linearly-polarized light in the plane of the QW, and investigate the possible observation of FIR features which could be unambiguously associated to *lh* cyclotron resonances and *lh* intraexcitonic transitions.

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